

FORM PTO-1500 (Modified)
(REV 11-2000)

U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE

ATTORNEY'S DOCKET NUMBER

TRANSMITTAL LETTER TO THE UNITED STATES
DESIGNATED/ELECTED OFFICE (DO/EO/US)
CONCERNING A FILING UNDER 35 U.S.C. 371

100718- 354/ Beiersdorf 750-KGB

U.S. APPLICATION NO. (IF KNOWN, SEE 37 CFR

09/980388

INTERNATIONAL APPLICATION NO.

PCT/EP00/04938

INTERNATIONAL FILING DATE

30 May 2000 (30.05.00)

PRIORITY DATE CLAIMED

06 June 1999 (06.06.99)

TITLE OF INVENTION

Use of ascorbic acid and one more flavone derivatives and/or flavanone derivatives, in particular flavonoids for the preparation of cosmetic or dermatological preparations for the prophylaxis or alleviation of sunburn

APPLICANT(S) FOR DO/EO/US

Inge KRUSE, Frank RIPPKE, and Uwe SCHONROCK

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. ☒ This is an express request to begin national examination procedures (35 U.S.C. 371(f)). The submission must include items (5), (6), (9) and (24) indicated below.
4. ☒ The US has been elected by the expiration of 19 months from the priority date (Article 31).
5. ☒ A copy of the International Application as filed (35 U.S.C. 371 (c) (2))
 - a. ☐ is attached hereto (required only if not communicated by the International Bureau).
 - b. ☒ has been communicated by the International Bureau.
 - c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
6. ☒ An English language translation of the International Application as filed (35 U.S.C. 371(c)(2)).
 - a. ☒ is attached hereto.
 - b. ☐ has been previously submitted under 35 U.S.C. 154(d)(4).
7. ☐ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371 (c)(3))
 - a. ☐ are attached hereto (required only if not communicated by the International Bureau).
 - b. ☐ have been communicated by the International Bureau.
 - c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
 - d. ☐ have not been made and will not be made.
8. ☐ An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
9. ☐ An oath or declaration of the inventor(s) (35 U.S.C. 371 (c)(4)).
10. ☐ An English language translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371 (c)(5)).
11. ☐ A copy of the International Preliminary Examination Report (PCT/IPEA/409).
12. ☒ A copy of the International Search Report (PCT/ISA/210).

Items 13 to 20 below concern document(s) or information included:

13. ☒ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
14. ☐ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
15. ☒ A **FIRST** preliminary amendment.
16. ☐ A **SECOND** or **SUBSEQUENT** preliminary amendment.
17. ☐ A substitute specification.
18. ☐ A change of power of attorney and/or address letter.
19. ☐ A computer-readable form of the sequence listing in accordance with PCT Rule 13ter.2 and 35 U.S.C. 1.821 - 1.825.
20. ☐ A second copy of the published international application under 35 U.S.C. 154(d)(4).
21. ☐ A second copy of the English language translation of the international application under 35 U.S.C. 154(d)(4).
22. ☒ Certificate of Mailing by Express Mail
23. ☒ Other items or information:

Copy of WO 00/74641 A1

U.S. APPLICATION NO. (IF KNOWN, SEE 17 CFR

09/980388

INTERNATIONAL APPLICATION NO.

PCT/EP00/04938

ATTORNEY'S DOCKET NUMBER

100718-354/ Beiersdorf 7

24. The following fees are submitted:

BASIC NATIONAL FEE (37 CFR 1.492 (a) (1) - (5)) :

- ☐ Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO \$1040.00
- ☒ International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO \$890.00
- ☐ International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO \$740.00
- ☐ International preliminary examination fee (37 CFR 1.482) paid to USPTO but all claims did not satisfy provisions of PCT Article 33(1)-(4) \$710.00
- ☐ International preliminary examination fee (37 CFR 1.482) paid to USPTO and all claims satisfied provisions of PCT Article 33(1)-(4) \$100.00

ENTER APPROPRIATE BASIC FEE AMOUNT =

\$890.00

Surcharge of \$130.00 for furnishing the oath or declaration later than months from the earliest claimed priority date (37 CFR 1.492 (e))

☐ 20 ☐ 30

\$0.00

CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE	
Total claims	10 - 20 =	0	x \$18.00	\$0.00
Independent claims	1 - 3 =	0	x \$84.00	\$0.00
Multiple Dependent Claims (check if applicable)			<input type="checkbox"/>	\$0.00

TOTAL OF ABOVE CALCULATIONS =

\$890.00

☐ Applicant claims small entity status. See 37 CFR 1.27. The fees indicated above are reduced by 1/2.

\$0.00

SUBTOTAL =

\$890.00

Processing fee of \$130.00 for furnishing the English translation later than months from the earliest claimed priority date (37 CFR 1.492 (f))

☐ 20 ☐ 30

\$0.00

TOTAL NATIONAL FEE =

\$890.00

Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31) (check if applicable).

☐

\$0.00

TOTAL FEES ENCLOSED =

\$890.00

Amount to be:
refunded \$
charged \$

- a. ☐ A check in the amount of _____ to cover the above fees is enclosed.
- b. ☒ Please charge my Deposit Account No. 14-1263 in the amount of \$890.00 to cover the above fees. A duplicate copy of this sheet is enclosed.
- c. ☒ The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. 14-1263. A duplicate copy of this sheet is enclosed.
- d. ☐ Fees are to be charged to a credit card. **WARNING:** Information on this form may become public. Credit card information should not be included on this form. Provide credit card information and authorization on PTO-2038.

NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO:

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SIGNATURE

Kurt G. Briscoe

NAME

33,141

REGISTRATION NUMBER

11-30-2001

DATE

09/980388

JC10 REC'D PCT/PTO 30 NOV 2001

Express Mail Label No. EV 015940951 US

Date Mailed: November 30, 2001

Beiersdorf 750-KGB

6713-Wihd-99-61

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

APPLICANTS : INGE KRUSE et al
SERIAL NO. : TO BE ASSIGNED
FILED : HEREWITH
FOR : USE OF ASCORBIC ACID AND ONE MORE FLAVONE
DERIVATIVES AND/OR FLAVANONE DERIVATIVES, IN
PARTICULAR FLAVONOIDS FOR THE PREPARATION OF
COSMETIC OR DERMATOLOGICAL PREPARATIONS FOR THE
PROPHYLAXIS OR ALLEVIATION OF SUNBURN
ART UNIT : TO BE ASSIGNED
EXAMINER : TO BE ASSIGNED

November 29, 2001

Hon. Commissioner of Patents
Washington, D.C. 20231

PRELIMINARY AMENDMENT

SIR:

Prior to examination, please amend the above-identified application as follows:

IN THE SPECIFICATION:

Insert as the first paragraph of the specification the following new paragraph: -- This
application is a 371 of PCT/EP00/04938 filed on May 30, 2000.--

IN THE CLAIMS:

Please amend claim 1 as follows:

1. Method of using ascorbic acid and/or ascorbyl compounds and one or more flavone derivatives and/or flavanone derivatives, in particular flavonoids for the preparation of cosmetic or dermatological preparations for the prophylaxis or alleviation of sunburn.

REMARKS

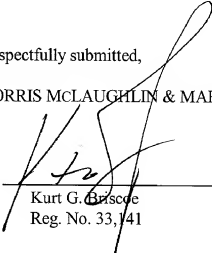
The amendment above convert the use claim to the more conventional method of use format, and otherwise place the claim in better form for U.S. examination.

Early and favorable action is earnestly solicited.

Respectfully submitted,

NORRIS MCLAUGHLIN & MARCUS, P.A.

By


Kurt G. Briscoe
Reg. No. 33,141

KGB:ja
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**MARK-UP SHOWING THE CHANGES MADE IN THE PREVIOUS CLAIM TO YIELD
THE CLAIM AS AMENDED ABOVE**

1. [The use of] **Method of using** ascorbic acid and/or ascorbyl compounds and one or more flavone derivatives and/or flavanone derivatives, in particular flavonoids for the preparation of cosmetic or dermatological preparations for the prophylaxis or alleviation of sunburn.

Beiersdorf Aktiengesellschaft
Hamburg

5

Description

Use of ascorbic acid and one more flavone derivatives and/or flavanone derivatives, in particular flavonoids for the preparation of cosmetic or dermatological preparations for the prophylaxis or alleviation of sunburn

10

The present invention relates to cosmetic and dermatological preparations.

The harmful effect of the ultraviolet part of solar radiation on the skin is generally known.

15

Depending on their respective wavelength, the rays have various effects on the skin as an organ: UV-C radiation with a wavelength below 290 nm is absorbed by the ozone layer in the earth's atmosphere and is therefore of no physiological significance. By contrast, rays in the range between 290 nm and 320 nm, the UV-B region, cause erythema, simple sunburn or even burns of greater or lesser severity.

20

Sunburn (*Erythema solare*) thus represents acute photodermatosis. It is a phototraumatic reaction in skin which has a sensitivity to light which is normal per se, as a result of an overdose of UV light. The symptoms of sunburn are skin reddening of greater or lesser severity and possible blistering. During the healing of sunburn, peeling of the areas of skin exposed to light is often observed.

25

The erythema activity maximum of sunlight is stated as the relatively narrow region around 308 nm.

30

For a long time it has incorrectly been assumed that the long-wavelength UV-A radiation with a wavelength between 320 nm and 400 nm has only a negligible biological effect and that, correspondingly, the UV-B rays are responsible for most photodamage to the human skin. However, in the meantime, numerous studies have demonstrated that UV-A radiation is far more hazardous than UV-B radiation with regard to the triggering of

photodynamic, specifically phototoxic, reactions and chronic changes in the skin. The harmful effect of UV-B radiation can also be further intensified by UV-A radiation.

5 Approximately 90% of the ultraviolet radiation which reaches the earth consists of UV-A rays. Whereas UV-B radiation varies greatly depending on numerous factors (e.g. season and time of day or latitude), UV-A radiation remains relatively constant from day to day irrespective of seasonal and diurnal or geographic factors. At the same time, most of the UV-A radiation penetrates into the living epidermis, while approximately 70% of the UV-B rays are retained by the horny layer.

10

Sunbathing is perceived by the majority of people as pleasant, and the disadvantageous consequences are not initially taken into consideration. However, in recent years, knowledge about the negative effects of excessively intensive solar irradiation has emerged, for which reason greater amounts and more strongly protecting sunscreens are used. To protect against UV-B radiation, as a prophylaxis for sunburn, numerous compounds are known, most of which are derivatives of 3-benzylidenecamphor, of 4-aminobenzoic acid, of cinnamic acid, of salicylic acid, of benzophenone and also of 2-phenylbenzimidazole.

15

20 The light absorption behavior of light protection filter substances is generally very well known and documented, especially since most industrialized countries have positive lists for the use of such substances, which impose very strict standards on the documentation. Depending on which range of UV light is absorbed, a differentiation is made between UV-B filters, UV-A filters and broadband filters (which demonstrate a filter action over the UV-B filters, UV-A filters and broadband filters (which demonstrate a filter action over the entire UV-A and UV-B range). Appropriate choice of the UV filter and its concentration in sunscreen compositions makes it possible to influence the degree of shielding from UV light. For the concentration of the substances in the finished formulations, the absorbance values can at best be a guide since interaction with ingredients of the formulation or of the skin itself may result in imponderables. In addition, it is usually difficult to estimate beforehand how uniformly and thickly the filter substance is distributed in and on the horny layer of the skin.

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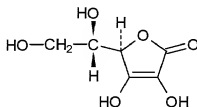
An effective prophylaxis of sunburn has hitherto only been achieved through the use of classic UV filters or UV blockers. Effective alleviation of already manifest sunburn was hitherto difficult.

35

The aim therefore was to remedy the shortcomings of the prior art.

The use of ascorbic acid in cosmetics and dermatological preparations is known per se.

- 5 L-ascorbic acid {(R)-5-[(S)-1,2-dihydroxyethyl]-3,4-dihydroxy-5-H-furan-2-one, vitamin C} is characterized by the structural formula

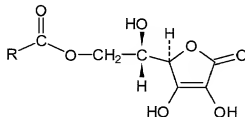


- 10 It is virtually soluble in water, relatively soluble in alcohol, insoluble in ether, petroleum ether, chloroform, benzene and in fats and fatty oils. Ascorbic acid is an enediol and, being a reductone, has a strong reducing action. Ascorbic acid is heat-sensitive and, particularly in the presence of traces of heavy metals and in an alkaline medium, is decomposed by light and atmospheric oxygen, but in a pure, dry state is relatively stable
- 15 toward light, air and heat.

In cosmetic and dermatological preparations, ascorbyl compounds are often used instead of ascorbic acid, preferably ascorbyl esters of fatty acids, particularly preferably ascorbyl palmitate since the sensitivity of these compounds to oxidative influence is greatly

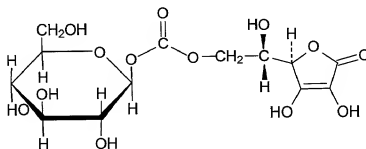
20 reduced compared with ascorbic acid and in most cases these compounds have better solubility in oil, which may offer galenical advantages.

Ascorbyl compounds in the narrower sense are, in particular, the ascorbyl esters of the general structure

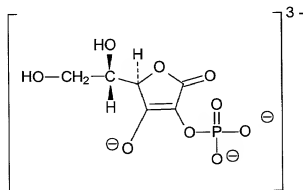


where R may be a branched or unbranched alkyl radical having up to 25 carbon atoms.

Ascorbyl glycosides are also advantageous for the purposes of the present invention, in particular ascorbyl glucosides, in particular ascorbyl 2-glucoside, which is represented by the structure



Ascorbyl compounds in the narrower sense are also ascorbyl phosphates, particularly advantageously the ascorbyl 2-phosphates of ascorbic acid or the alkali metal, alkaline earth metal and zinc salts thereof, or mixed salts of such cations.



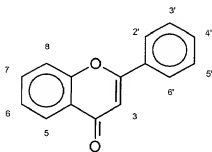
Shown above is a triple-deprotonated ascorbyl phosphate ion, although other deprotonation stages are also advantageous for the purposes of the present invention.

Preference is given to the sodium, magnesium and zinc salts, for example sodium ascorbyl phosphate.

The use of flavones and flavonoids in cosmetics and dermatology is known per se. For example, DE-A 44 44 238 describes combinations of cinnamic acid derivatives and flavone glycosides, for example α -glycosylrutin as antioxidants and as active ingredients against other indications.

- It was therefore surprising and could not have been foreseen by the person skilled in the art that the use of ascorbic acid and one or more flavone derivatives and/or flavanone derivatives, in particular flavonoids for the preparation of cosmetic or dermatological preparations for the prophylaxis or alleviation of sunburn would remedy the disadvantages of the prior art.

Flavone and its derivatives (often also collectively called "flavones") are characterized by the following basic structure (substitution positions are given):



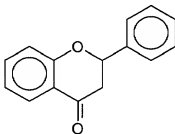
Some of the more important flavones, which can also be found in living nature, are given in the table below:

	OH substitution positions							
	3	5	7	8	2'	3'	4'	5'
Flavone	-	-	-	-	-	-	-	-
Flavonol	+	-	-	-	-	-	-	-
Chrysin	-	+	+	-	-	-	-	-
Galangin	+	+	+	-	-	-	-	-
Apigenin	-	+	+	-	-	-	+	-
Fisetin	+	-	+	-	-	+	+	-
Luteolin	-	+	+	-	-	+	+	-
Kaempferol	+	+	+	-	-	-	+	-
Quercetin	+	+	+	-	-	+	+	-
Morin	+	+	+	-	+	-	+	-
Robinetin	+	-	+	-	-	+	+	+
Gossypetin	+	+	+	+	-	+	+	-
Myricetin	+	+	+	-	-	+	+	+

In nature, flavones are usually in glycosylated form.

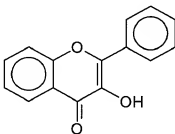
Flavonoids are glycosides of flavones, of flavanones, the basic skeleton of which is characterized by the following structure:

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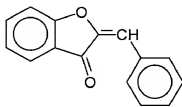


of 3-hydroxyflavones (flavonols), the basic skeleton of which is characterized by the following structure:

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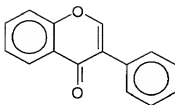


of aurones, the basic skeleton of which is characterized by the following structure:



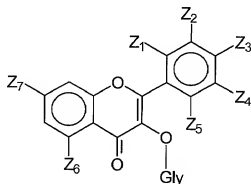
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and also of isoflavones, the basic skeleton of which is characterized by the following structure:



According to the invention, the flavonoids are preferably chosen from the group of substances having the generic structural formula

5

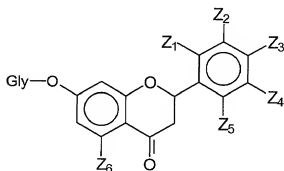


where Z_1 - Z_8 independently of one another are chosen from the group consisting of H, OH, alkoxy and hydroxyalkoxy, where the alkoxy or hydroxyalkoxy groups may be branched or unbranched and may have 1-18 carbon atoms, and where Gly is chosen from the group of mono- and oligoglycoside radicals.

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According to the invention, the flavonoids can however also be advantageously chosen from the group of substances of the generic structural formula

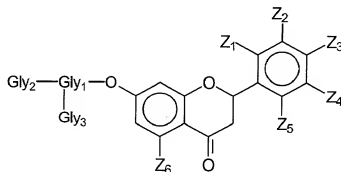
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where Z_1 - Z_6 independently of one another are chosen from the group consisting of H, OH, alkoxy and hydroxyalkoxy, where the alkoxy or hydroxyalkoxy groups can be branched or unbranched and may have 1-18 carbon atoms, and where Gly is chosen from the group consisting of mono- and oligoglycoside radicals.

5

Such structures may preferably be chosen from the group of substances of the generic structural formula



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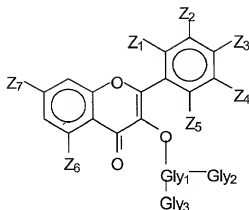
where Gly_1 , Gly_2 and Gly_3 independently of one another are monoglycoside radicals. Gly_2 and Gly_3 may also, individually or together, represent saturations by hydrogen atoms.

15

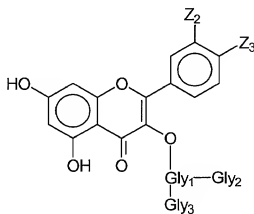
Gly_1 , Gly_2 and Gly_3 independently of one another are preferably chosen from the group consisting of hexosyl radicals, in particular rhamnosyl radicals and glucosyl radicals. However, if desired, it is also advantageous to use other hexosyl radicals, for example allosyl, altrosyl, galactosyl, gulosyl, idosyl, mannosyl and talosyl. It may also be advantageous according to the invention to use pentosyl radicals.

20

Z_1 - Z_6 independently of one another are advantageously chosen from the group consisting of H, OH, methoxy, ethoxy and 2-hydroxyethoxy, and the flavone glycosides have the structure



The flavone glycosides according to the invention are particularly advantageously chosen from the group represented by the following structure:



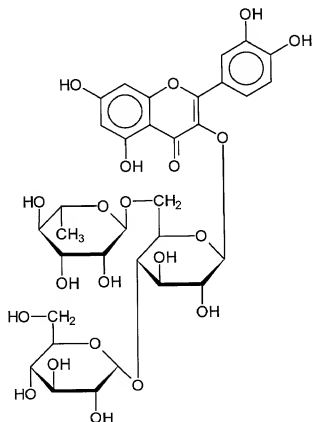
where Gly₁, Gly₂ and Gly₃ independently of one another are monoglycoside radicals. Gly₂ and Gly₃ can also, individually or together, represent saturations by hydrogen atoms.

- 10 Gly₁, Gly₂ and Gly₃ independently of one another are preferably chosen from the group consisting of hexosyl radicals, in particular rhamnosyl radicals and glucosyl radicals. However, if desired, it is also advantageous to use other hexosyl radicals, for example allosyl, altrosyl, galactosyl, gulosyl, idosyl, mannosyl and talosyl. It may also be advantageous according to the invention to use pentosyl radicals.

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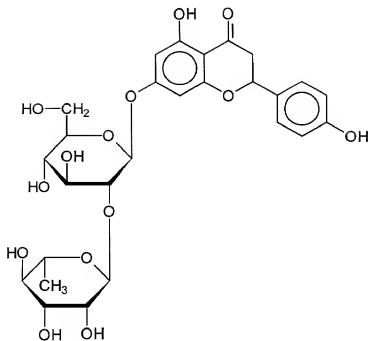
For the purposes of the present invention, it is particularly advantageous to choose the flavone glycoside(s) from the group consisting of α -glucosylrutin, α -glucosylmyricitrin, α -glucosylisoquercitrin and α -glucosylquercitrin.

One flavonoid which is particularly advantageous according to the invention is α -glucosylrutin. It is characterized by the following structure:



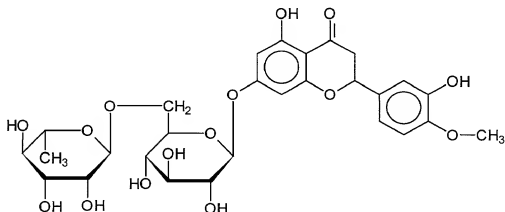
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Another particularly advantageous flavonoid according to the invention is naringin (aurantiin, naringenin 7-rhamnoglucoiside). It is characterized by the following structure:



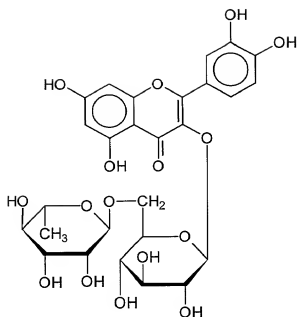
Another particularly advantageous flavonoid according to the invention is hesperidin (3',5,7-trihydroxy-4'-methoxyflavanone-7-rutinoside, hesperidoside, hesperetin-

5 7-O-rutinoside). It is characterized by the following structure:



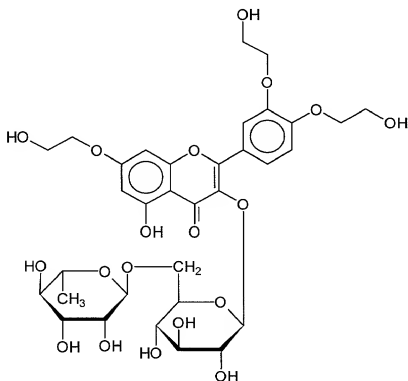
Another particularly advantageous flavonoid according to the invention is rutin (3,3',4',5,7-pentahydroxyflavone-3-rutinoside, quercetin-3-rutinoside, sophorin, birutan, rutabion, taurutin, phytomelin, melin). It is characterized by the following structure:

10

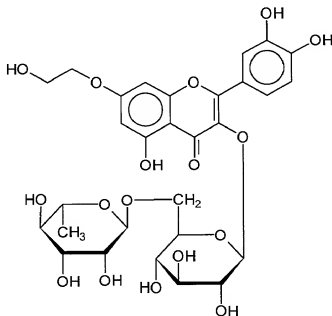


Another particularly advantageous flavonoid according to the invention is troxerutin (3,5-dihydroxy-3',4',7-tris(2-hydroxyethoxy)flavone-3-(6-O-(6-deoxy-α-L-mannopyranosyl)-

5 β-D-glucopyranoside)). It is characterized by the following structure:

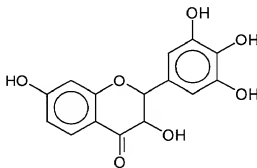


Another particularly advantageous flavonoid according to the invention is monoxerutin (3,3',4',5-tetrahydroxy-7-(2-hydroxyethoxy)flavone-3-(6-O-(6-deoxy- α -L-mannopyranosyl)- β -D-glucopyranoside)). It is characterized by the following structure:



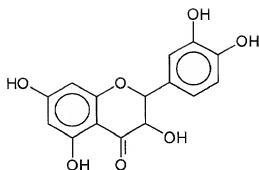
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Another particularly advantageous flavonoid according to the invention is dihydrorobinetin (3,3',4',5',7-pentahydroxyflavanone). It is characterized by the following structure:

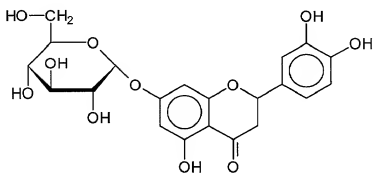


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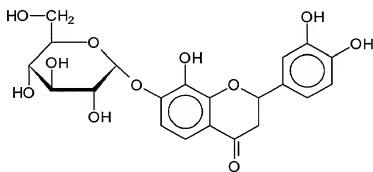
Another particularly advantageous flavonoid according to the invention is taxifolin (3,3',4',5',7-pentahydroxyflavanone). It is characterized by the following structure:



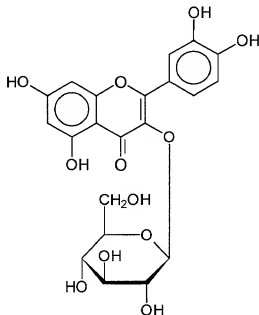
Another particularly advantageous flavonoid according to the invention is eriodictyol-7-glucoside (3',4',5,7-tetrahydroxyflavanone-7-glucoside). It is characterized by the following structure:



Another particularly advantageous flavonoid according to the invention is flavanomarein (3',4',7,8-tetrahydroxyflavanone-7-glucoside). It is characterized by the following structure:



Another particularly advantageous flavonoid according to the invention is isoquercitrin (3,3',4',5,7-pentahydroxyflavanone-3-(β -D-glucopyranoside). It is characterized by the following structure:



According to the invention, the flavone derivative(s) and/or flavanone derivative(s), in particular flavonoids, are advantageously present in cosmetic or dermatological preparations preferably in amounts of from 0.001% by weight to 10% by weight, preferably in amounts from 0.05% by weight to 5% by weight, particularly preferably in amounts of 0.1-2.0% by weight, based on the total weight of the preparations.

According to the invention, the ascorbyl compound or the ascorbyl compounds, in particular vitamin C, is/are advantageously present in cosmetic or dermatological preparations preferably in amounts of from 0.001% by weight to 10% by weight, preferably in amounts of from 0.05% by weight to 5% by weight, particularly preferably in amounts of 0.1-2.0% by weight, based on the total weight of the preparations.

The novel combination of at least one flavone derivative and/or flavanone derivative, in particular at least one flavonoid and at least one ascorbyl compound, in particular vitamin C, is, for the purposes of this specification, also collectively referred to as "active ingredient according to the invention" or "active ingredient used according to the

invention" or "active ingredient combination used according to the invention" or given synonymous designations.

The specification JP-A Hei-06-138,941 describes oral preparations containing water-soluble glycosides which can be chosen, for example, from the group consisting of α -glucosylrutin, α -glucosylmyricetin, α -glucosylisoquercitrin and α -glucosylquercitrin. The specification JP-A Hei-04-363,395 describes a method of preventing the decomposition of perfume constituents, which is characterized inter alia by an addition of α -glucosylrutin to the corresponding preparations. In addition, the specifications EP-A 586 303 and EP-A 595 694 describe the use of flavonoids as antioxidants or light protection substances in cosmetics.

However, these specifications do not contain any information which could point in the direction of the present invention.

It was rather surprising and could not have been foreseen by the person skilled in the art that preparations according to the use according to the invention would ensure excellent prophylaxis for sunburn, even if the additional presence of UV filter substances or UV blockers is dispensed with. It was also surprising that by following the teaching according to the invention, a significant alleviation of already manifest sunburn is observed.

The cosmetic or dermatological preparations according to the invention can have the customary composition and can be used for the treatment, care and cleansing of the skin. They preferably comprise from 0.001% by weight to 10% by weight, preferably from 0.05% by weight to 5% by weight, in particular 0.1-2.0% by weight, based on the total weight of the preparations, of active ingredient combinations used according to the invention.

According to the invention, it is preferred to add complexing agents to the active ingredient combinations used according to the invention or to cosmetic or dermatological preparations comprising such active ingredient combinations.

Complexing agents are auxiliaries used in cosmetics or medicinal pharmaceutical technology which are known per se. By complexing undesired metals such as Mn, Fe, Cu and others, it is possible, for example, to prevent undesired chemical reactions in cosmetic or dermatological preparations.

Complexing agents, in particular chelating agents, form complexes with metal atoms, which, in the presence of one or more polybasic complexing agents, i.e. chelating agents, represent metallacycles. Chelates are compounds in which a single ligand occupies more than one co-ordination site on a central atom. In this case, compounds which are normally extended are thus closed as a result of complex formation via a metal atom or a metal ion to form rings. The number of bonded ligands depends on the co-ordination number of the central metal. A prerequisite for formation of the chelate is that the compound reacting with the metal contains two or more atomic groupings which act as electron donors.

The complexing agent(s) can advantageously be chosen from the group of customary compounds, preferably at least one substance from the group consisting of tartaric acid and anions thereof, citric acid and anions thereof, aminopolycarboxylic acids and anions thereof (such as, for example, ethylenediaminetetraacetic acid (EDTA) and anions thereof, nitrilotriacetic acid (NTA) and anions thereof, hydroxyethylenediaminetriacetic acid (HOEDTA) and anions thereof, diethyleneaminopentaacetic acid (DPTA) and anions thereof, trans-1,2-diaminocyclohexanetetraacetic acid (CDTA) and anions thereof).

According to the invention, the complexing agent(s) is/are advantageously present in cosmetic or dermatological preparations preferably in amounts from 0.01% by weight to 10% by weight, preferably in amounts from 0.05% by weight to 5% by weight, particularly preferably in amounts of 0.1-2.0% by weight, based on the total weight of the preparations.

For use, according to the invention, the cosmetic and dermatological preparations are applied to the skin and/or the hair in an adequate amount in the manner customary for cosmetics.

Cosmetic and dermatological preparations according to the invention can be in various forms. Thus, they can, for example, be a solution, an anhydrous preparation, an emulsion or microemulsion of the water-in-oil (W/O) type or of the oil-in-water (O/W) type, a multiple emulsion, for example of the water-in-oil-in-water (W/O/W) type, a gel, a solid stick, an ointment or else an aerosol. It is also advantageous to administer isoquercitrin in encapsulated form, for example in collagen matrices and other customary encapsulation materials, for example as cellulose encapsulations, in gelatin, wax matrices or

liposomally encapsulated. In particular, wax matrices, as are described in DE A 43 08 282, have proven to be favorable.

The cosmetic and dermatological preparations according to the invention, can comprise
 5 cosmetic auxiliaries as are usually used in such preparations, for example preservatives, bactericides, perfumes, antifoams, dyes, pigments which have a coloring action, thickeners, surfactants, emulsifiers, emollients, moisturizers and/or humectants, fats, oils, waxes or other customary constituents of a cosmetic or dermatological preparation, such
 10 as alcohols, polyols, polymers, foam stabilizers, electrolytes, organic solvents or silicone derivatives.

In particular, active ingredient combinations used according to the invention can also be combined with other antioxidants and/or free-radical scavengers.

15 Such antioxidants are advantageously chosen from the group consisting of amino acids (for example glycine, histidine, tyrosine, tryptophan) and derivatives thereof, imidazoles (for example urocanic acid) and derivatives thereof, peptides such as D,L-carnosine, D-carnosine, L-carnosine and derivatives thereof (for example anserine), carotenoids, carotenes (for example α -carotene, β -carotene, lycopene) and derivatives thereof,
 20 chlorogenic acid and derivatives thereof, lipoic acid and derivatives thereof (for example dihydrolipoic acid), aurothioglucose, propylthiouracil and other thiols (for example thioredoxin, glutathione, cysteine, cystine, cystamine and the glycosyl, N-acetyl, methyl, ethyl, propyl, amyl, butyl and lauryl, palmitoyl, oleyl, γ -linoleyl, cholesteryl and glyceryl esters thereof) and salts thereof, dilauryl thiodipropionate, distearyl thiodipropionate,
 25 thiodipropionic acid and derivatives thereof (esters, ethers, peptides, lipids, nucleotides, nucleosides and salts) and sulfoximine compounds (for example buthionine sulfoximines, homocysteine sulfoximine, buthionine sulfones, penta-, hexa- and heptathionine sulfoximine) in very low tolerated doses (for example pmol to μ mol/kg), and furthermore (metal) chelating agents (for example α -hydroxy-fatty acids, palmitic acid, phytic acid, lactoferrin), α -hydroxy acids (for example citric acid, lactic acid, malic acid), humic acid,
 30 bile acid, bile extracts, bilirubin, biliverdin, EDTA, EGTA and derivatives thereof, unsaturated fatty acids and derivatives thereof (for example γ -linolenic acid, linoleic acid, oleic acid), folic acid and derivatives thereof, ubiquinone and ubiquinol and derivatives thereof, tocopherols and derivatives (for example vitamin E acetate), vitamin A and
 35 derivatives (vitamin A palmitate) and coniferyl benzoate of benzoin resin, rutinic acid and derivatives thereof, butyl hydroxytoluene, butyl hydroxyanisole, nordihydroguaiacic acid,

nordihydroguaiaretic acid, trihydroxybutyrophenone, uric acid and derivatives thereof, mannose and derivatives thereof, sesamol, sesamolin, zinc and derivatives thereof (for example ZnO , ZnSO_4), selenium and derivatives thereof (for example selenium methionine), stilbenes and derivatives thereof (for example stilbene oxide, trans-stilbene oxide) and the derivatives of said active ingredients which are suitable according to the invention (salts, esters, ethers, sugars, nucleotides, nucleosides, peptides and lipids).

The amount of the abovementioned antioxidants (one or more compounds) in the preparations is preferably from 0.001 to 30% by weight, particularly preferably 0.05-20% by weight, in particular 1-10% by weight, based on the total weight of the preparation.

If vitamin E and/or derivatives thereof is or are the additional antioxidant(s), it is advantageous to choose the respective concentrations thereof from the range 0.001-10% by weight, based on the total weight of the preparation.

If vitamin A or vitamin A derivatives or carotenes or derivatives thereof is or are the additional antioxidant(s), it is advantageous to choose the respective concentrations thereof from the range 0.001-10% by weight, based on the total weight of the preparation.

Emulsions according to the invention are advantageous and comprise, for example, said fats, oils, waxes and other fatty substances, and also water and an emulsifier, as is customarily used for this type of formulation.

The lipid phase can advantageously be chosen from the following group of substances:

- mineral oils, mineral waxes;
- oils, such as triglycerides of capric or of caprylic acid, also natural oils such as, for example, castor oil;
- fats, waxes and other natural and synthetic fatty substances, preferably esters of fatty acids with alcohols of low carbon number, for example with isopropanol, propylene glycol or glycerol, or esters of fatty alcohols with alkanolic acids of low carbon number or with fatty acids;
- alkyl benzoates;
- silicone oils, such as dimethylpolysiloxanes, diethylpolysiloxanes, diphenylpolysiloxanes and mixed forms thereof.

For the purposes of the present invention, the oil phase of the emulsions, oleogels and hydrodispersions or lipodispersions is advantageously chosen from the group of esters of saturated and/or unsaturated, branched and/or unbranched alkanecarboxylic acids having a chain length of from 3 to 30 carbon atoms and saturated and/or unsaturated, branched and/or unbranched alcohols having a chain length of from 3 to 30 carbon atoms, from the group of esters of aromatic carboxylic acids and saturated and/or unsaturated, branched and/or unbranched alcohols having a chain length of from 3 to 30 carbon atoms. Such ester oils can then be advantageously chosen from the group consisting of isopropyl myristate, isopropyl palmitate, isopropyl stearate, isopropyl oleate, n-butyl stearate, n-hexyl laurate, n-decyl oleate, isooctyl stearate, isononyl stearate, isononyl isononanoate, 2-ethylhexyl palmitate, 2-ethylhexyl laurate, 2-hexyldodecyl stearate, 2-octyldodecyl palmitate, oleyl oleate, oleyl erucate, erucyl oleate, erucyl erucate and synthetic, semi-synthetic and natural mixtures of such esters, e.g. jojoba oil.

The oil phase can also advantageously be chosen from the group of branched and unbranched hydrocarbons and hydrocarbon waxes, silicone oils, dialkyl ethers, from the group of saturated or unsaturated, branched or unbranched alcohols, and also fatty acid triglycerides, namely the triglycerol esters of saturated and/or unsaturated, branched and/or unbranched alkanecarboxylic acids having a chain length of from 8 to 24, in particular 12-18, carbon atoms. The fatty acid triglycerides can advantageously be chosen, for example, from the group of synthetic, semi-synthetic and natural oils, e.g. olive oil, sunflower oil, soybean oil, groundnut oil, rapeseed oil, almond oil, palm oil, coconut oil, palm kernel oil and the like.

For the purposes of the present invention, any mixtures of such oil and wax components can also advantageously be used. When required, it may also be advantageous to use waxes, for example cetyl palmitate, as the sole lipid component of the oil phase.

The oil phase is advantageously chosen from the group consisting of 2-ethylhexyl isostearate, octyldodecanol, isotridecyl isononanoate, isoeicosane, 2-ethylhexyl cocoate, C_{12} - C_{15} -alkyl benzoate, caprylic/capric triglyceride and dicaprylyl ether.

Mixtures of C_{12} - C_{15} -alkyl benzoate and 2-ethylhexyl isostearate, mixtures of C_{12} - C_{15} -alkyl benzoate and isotridecyl isononanoate and mixtures of C_{12} - C_{15} -alkyl benzoate, 2-ethylhexyl isostearate and isotridecyl isononanoate are particularly advantageous.

For the purposes of the present invention, of the hydrocarbons, paraffin oil, squalane and squalene can advantageously be used.

5 The oil phase can advantageously also contain cyclic or linear silicone oils or can consist entirely of such oils, although it is preferable to use an additional content of other oil phase components in addition to the silicone oil or silicone oils.

10 Cyclomethicone (octamethylcyclotetrasiloxane) is advantageously used as the silicone oil to be used according to the invention. However, other silicone oils can also be advantageously used for the purposes of the present invention, for example hexamethylcyclotrisiloxane, polydimethylsiloxane, poly(methylphenylsiloxane).

15 Mixtures of cyclomethicone and isotridecyl isononanoate and mixtures of cyclomethicone and 2-ethylhexyl isostearate are particularly advantageous.

20 If appropriate, the aqueous phase of the preparations according to the invention advantageously comprises alcohols, diols or polyols of low carbon number and ethers thereof, preferably ethanol, isopropanol, propylene glycol, glycerol, ethylene glycol, ethylene glycol monoethyl or monobutyl ether, propylene glycol monomethyl, monoethyl or monobutyl ether, diethylene glycol monomethyl or monoethyl ether and analogous products, also alcohols of low carbon number, for example ethanol, isopropanol, 1,2-propanediol and glycerol, and, in particular, one or more thickeners, which can advantageously be chosen from the group consisting of silicon dioxide, aluminum silicates, polysaccharides and derivatives thereof, for example hyaluronic acid, xanthan gum and hydroxypropylmethylcellulose, particularly advantageously from the group consisting of polyacrylates, preferably a polyacrylate from the group consisting of Carbopols, for example Carbopols of types 980, 981, 1382, 2984 and 5984, in each case individually or in combination.

30 In particular, mixtures of the abovementioned solvents are used. In the case of alcoholic solvents, water may be a further constituent.

35 Emulsions according to the invention are advantageous and comprise, for example, said fats, oils, waxes and other fatty substances, and also water and an emulsifier, as is customarily used for this type of formulation.

Gels according to the invention customarily comprise alcohols of low carbon number, for example ethanol, isopropanol, 1,2-propanediol, glycerol, and water and/or an abovementioned oil in the presence of a thickener which, in the case of oily-alcoholic gels, is preferably silicon dioxide or an aluminum silicate, and in the case of aqueous-alcoholic or alcoholic gels, is preferably a polyacrylate.

Suitable propellants for preparations according to the invention which can be sprayed from aerosol containers are the customary known, readily volatile, liquefied propellants, for example hydrocarbons (propane, butane, isobutane), which may be used alone or in mixtures with one another. Compressed air can also be used advantageously.

Preparations according to the invention can advantageously also comprise substances which absorb UV radiation in the UVB region, the total amount of filter substances being, for example, 0.1% by weight to 30% by weight, preferably 0.5 to 10% by weight, in particular 1.0 to 6.0% by weight, based on the total weight of the preparations, in order to provide cosmetic formulations which protect the hair or skin from the entire range of ultraviolet radiation. They can also be used as sunscreen compositions for hair or skin.

If the preparations according to the invention comprise UVB filter substances, these may be oil-soluble or water-soluble. Advantageous oil-soluble UVB filter substances are, for example:

- 3-benzylidenecamphor derivatives, preferably 3-(4-methylbenzylidene)camphor and 3-benzylidenecamphor;
- 4-aminobenzoic acid derivatives, preferably 2-ethylhexyl 4-(dimethylamino)benzoate and amyl 4-(dimethylamino)benzoate;
- esters of cinnamic acid, preferably 2-ethylhexyl 4-methoxycinnamate and isopentyl 4-methoxycinnamate;
- esters of salicylic acid, preferably 2-ethylhexyl salicylate, 4-isopropylbenzyl salicylate and homomenthyl salicylate,
- derivatives of benzophenone, preferably 2-hydroxy-4-methoxybenzophenone, 2-hydroxy-4-methoxy-4'-methylbenzophenone and 2,2'-dihydroxy-4-methoxybenzophenone;
- esters of benzalmalonic acid, preferably di(2-ethylhexyl) 4-methoxybenzalmalonate and
- 2,4,6-tris(p-2-ethylhexoxy carbonylanilino)-1,3,5-triazine.

Advantageous water-soluble UVB filters are, for example:

- salts of 2-phenylbenzimidazole-5-sulfonic acid, such as its sodium, potassium or its triethanolammonium salt, and the sulfonic acid itself;
- sulfonic acid derivatives of benzophenones, preferably 2-hydroxy-4-methoxy-
5 benzophenone-5-sulfonic acid and its salts;
- sulfonic acid derivatives of 3-benzylidenecamphor, such as, for example, 4-(2-oxo-3-bornylidenemethyl)benzenesulfonic acid, 2-methyl-5-(2-oxo-3-bornylidenemethyl)sulfonic acid and their salts, and also 1,4-di(2-oxo-10-sulfo-
10 3-bornylidenemethyl)benzene and its salts (the corresponding 10-sulfato compounds, for example the corresponding sodium, potassium or triethanolammonium salt) also referred to as benzene-1,4-di(2-oxo-3-bornylidenemethyl)-10-sulfonic acid.

The list of said UVB filters which can be used in combination with the active ingredient
15 combinations according to the invention is not of course intended to be limiting.

The invention also provides for the use of a combination of the active ingredient combinations used according to the invention with at least one UVB filter as an antioxidant and for the use of a combination of the active ingredient combinations used
20 according to the invention with at least one UVB filter as an antioxidant in a cosmetic or dermatological preparation.

It may also be advantageous to combine the active ingredient combinations used according to the invention with UVA filters which have to date customarily been present
25 in cosmetic preparations. These substances are preferably derivatives of dibenzoyl-methane, in particular 1-(4'-tert-butylphenyl)-3-(4'-methoxyphenyl)propane-1,3-dione and 1-phenyl-3-(4'-isopropylphenyl)propane-1,3-dione. These combinations and preparations comprising these combinations are also provided by the invention. The amounts which may be used are as for the UVB combination.

Cosmetic and dermatological preparations having an effective content of active ingredient combinations according to the invention can also comprise inorganic pigments which are normally used in cosmetics for protecting the skin against UV rays. These are oxides of titanium, zinc, zirconium, silicon, manganese, cerium and mixtures thereof, and
35 modifications in which the oxides are the active agents. Particular preference is given to pigments based on titanium dioxide.

These combinations of UVA filter and pigment and preparations which comprise this combination are also provided by the invention. The quantities which may be used are as stated for the aforementioned combinations.

5

For the purposes of the present invention, electrolytes are understood as meaning water-soluble alkali metal, ammonium, alkaline earth metal (including magnesium) and zinc salts of inorganic anions and any mixtures of such salts, it being necessary to ensure that these salts are pharmaceutically or cosmetically safe.

10

The anions according to the invention are preferably chosen from the group consisting of chlorides, sulfates and hydrogensulfates, phosphates, hydrogenphosphates and linear and cyclic oligophosphates and carbonates and hydrogencarbonates.

15

These cosmetic or dermatological preparations can also be in the form of aerosols with the auxiliaries usually used for this purpose.

The amount of active ingredient compositions used according to the invention in these preparations is preferably 0.01-10% by weight, preferably 0.05-5% by weight, in particular 0.1-2.0% by weight, based on the total weight of the preparations.

20

The examples below serve to illustrate the present invention without limiting it. Unless stated otherwise, all quantities, proportions and percentages are by weight and based on the total amount or on the total weight of the preparations.

Example 1

O/W cream

	% by wt.
Glyceryl stearate	5.00
Cetyl alcohol	5.00
Isopropyl palmitate	7.00
Cyclomethicone	5.00
Ascorbic acid	3.00
α -Glucosylrutin	0.30
NaOH, 45% strength	1.00
Butylene glycol	3.00
$\text{Na}_2\text{H}_2\text{EDTA}$	0.20
Dyes, perfume, preservatives	q.s.
Water	ad 100.00

Example 2

5

O/W lotion

	% by wt.
Steareth-20	3.00
Cetyl alcohol	3.00
Cyclomethicone	6.00
Carbomer	0.60
$\text{Na}_2\text{H}_2\text{EDTA}$	0.20
Butylene glycol	3.00
NaOH, 45% strength	0.40
Ascorbic acid	0.50
α -Glucosylrutin	0.10
Dyes, perfume, preservatives	q.s.
Water	ad 100.00

Example 3

W/O cream

	% by wt.
Polyglyceryl-2 dipolyhydroxystearate	5.00
Caprylic/capric triglycerides	15.00
Butylene glycol	3.00
Na ₂ H ₂ EDTA	0.20
MgSO ₄	0.70
NaOH, 45% strength	0.32
Ascorbic acid	1.00
α-Glucosylrutin	0.20
Dyes, perfume, preservatives	q.s.
Water	ad 100.00

5 **Example 4**

O/W gel

	% by wt.
Xanthan gum	2.00
Butylene glycol	3.00
Na ₂ H ₂ EDTA	0.20
NaOH, 45% strength	0.32
Ascorbic acid	1.00
α-Glucosylrutin	0.20
Dyes, perfume, preservatives	q.s.
Water	ad 100.00

Example 5

O/W hair treatment

	% by wt.
Bis-diglyceryl polyacyladipate-2	3.00
Behenyl alcohol	4.00
Butylene glycol	3.00
Cetrimonium chloride	5.00
Citric acid	0.50
Na ₂ H ₂ EDTA	0.20
NaOH, 45% strength	0.16
Ascorbic acid	0.50
α -Glucosylrutin	0.10
Dyes, perfume, preservatives	q.s.
Water	ad 100.00

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Example 6

Sunscreen emulsion

	% by wt.
Cyclomethicone	2.00
Cetylstearyl alcohol + PEG 40 hydrogenated	2.50
castor oil + sodium cetylstearyl sulfate	
Glyceryl lanolate	1.00
Caprylic/capric triglyceride	0.10
Laurylmethicone copolyol	2.00
Octylstearate	3.00
Castor oil	4.00
Glycerol	3.00
Acrylamide/sodium acrylate copolymer	0.30
Hydroxypropylmethylcellulose	0.30
Octyl methoxycinnamate	5.00
Butylmethoxydibenzoylmethane	0.75
α -Glucosylrutin	0.50
Ascorbyl 2-glucoside	1.00
Epigallocatechin gallate	0.20
α -Tocopheryl acetate	1.00
Na ₃ HEDTA	1.50
Preservatives, dyes, perfume	q.s.
Water	ad 100.00

Example 7

O/W lotion

	% by wt.
Paraffin oil (DAB 9)	8.00
Isopropyl palmitate	3.00
Petrolatum	4.00
Cetylstearyl alcohol	2.00
PEG 40 castor oil	0.50
Sodium cetylstearyl sulfate	0.50
Sodium carbomer	0.40
α -Glucosylrutin	0.50
Glycerol	3.00
Ascorbyl palmitate	1.00
Octyl methoxycinnamate	5.00
Butylmethoxydibenzoylmethane	1.00
Preservatives, dyes, perfume	q.s.
Epigallocatechin gallate	0.05
Water	ad 100.00

Example 8

5

O/W cream

	% by wt.
Paraffin oil (German Pharmacopoeia)	7.00
Avocado oil	4.00
Glyceryl monostearate	2.00
α -Glucosylrutin	0.80
Mg Ascorbyl phosphate	0.50
Sodium lactate	3.00
Glycerol	3.00
Preservative, dyes, perfume	q.s.
Epigallocatechin gallate	3.00
Water	ad 100.00

Patent claims

1. The use of ascorbic acid and/or ascorbyl compounds and one or more flavone derivatives and/or flavanone derivatives, in particular flavonoids for the preparation of
5 cosmetic or dermatological preparations for the prophylaxis or alleviation of sunburn.

2. The use as claimed in claim 1, wherein the active ingredient(s) chosen from the group of flavones, flavanones and flavonoids is/are present in cosmetic or dermatological preparations in an effective content.
10

3. The use as claimed in claim 2, wherein the active ingredient(s) chosen from the group of flavones, flavanones and flavonoids is/are present in cosmetic or topical dermatological preparations in concentrations of 0.01-10% by weight, preferably 0.05-5% by weight, in particular 0.1-2.0% by weight, based on the total weight of the preparations.
15

4. The use as claimed in claim 1, wherein the active ingredient(s) chosen from the group of ascorbic acid and ascorbyl compounds is/are present in cosmetic or dermatological preparations in an effective amount.

20 5. The use as claimed in claim 1, wherein the ascorbyl compounds are chosen from the group ascorbyl palmitate, sodium ascorbyl phosphate, magnesium ascorbyl phosphate, zinc ascorbyl phosphate and ascorbyl 2-glucoside.

25 6. The use as claimed in claim 4, wherein the active ingredient(s) chosen from the group of ascorbic acid and ascorbyl compounds is/are present in cosmetic or topical dermatological preparations in concentrations of 0.001-10% by weight, preferably 0.05-5% by weight, in particular 0.1-2.0% by weight, based on the total weight of the preparations.

30 7. The use as claimed in claim 1, wherein the active ingredient chosen from the group of flavones, flavanones and flavonoids is α -glucosylrutin.

8. The use as claimed in claim 2, wherein the cosmetic or dermatological preparations additionally comprise one or more complexing agents.
35

9. The use as claimed in claim 7, wherein the complexing agent(s) is/are chosen from the group consisting of tartaric acid and anions thereof, citric acid and anions thereof, aminopolycarboxylic acids and anions thereof (such as, for example ethylenediaminetetraacetic acid and anions thereof, nitrilotriacetic acid and anions thereof, hydroxyethylenediaminotriacetic acid and anions thereof, diethylene-aminopentaacetic acid and anions thereof, trans-1,2-diaminocyclohexanetetraacetic acid and anions thereof).
10. The use as claimed in claim 7, wherein the complexing agent(s) is/are present in the cosmetic or dermatological preparation preferably in amounts of from 0.01% by weight to 10% by weight, preferably in amounts of from 0.05% by weight to 5% by weight, particularly preferably in amounts of 0.1-2.0% by weight, based on the total weight of the preparations.

Figure 6

bat-wi/99-061-oct.doc/ar

COMBINATION DECLARATION & POWER OF ATTORNEY

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name. I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled

USE OF ASCORBIC ACID AND ONE MORE FLAVONE DERIVATIVES AND/OR FLAVANONE
DERIVATIVES, IN PARTICULAR FLAVONOIDS FOR THE PREPARATION OF COSMETIC OR
DERMATOLOGICAL PREPARATIONS FOR THE PROPHYLAXIS OR ALLEVIATION OF SUNBURN

the specification of which was filed on ²⁰November 29, 2001

as Application Serial No. 09/980,388 and

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims.

I acknowledge the duty to disclose information which is material to the examination of this application in accordance with Title 37, Code of Federal Regulations §1.56(a).

I hereby claim foreign priority benefits under Title 35, United States Code, §119 of any foreign application(s) for patent or inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed:

Prior Foreign Application(s)

Priority Claimed

199 25 499.0 ✓
(Number)

Germany ✓
(Country)

⁰⁴
8 June 1999 ✓
(Day/Month/Yr. Filed)

X yes no

(Number)

(Country)

(Day/Month/Yr. Filed)

 yes no

I hereby claim the benefit under Title 35, United States Code, §120 of any United States application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of Title 35, United States Code, §112, I acknowledge the duty to disclose material information as defined in Title 37, Code of Federal Regulations, §1.56(a) which occurred between the filing date of the prior application and the national or PCT international filing date of this application:

(Application Serial No.)

(Filing Date)

(Status)
(patented,pending,abandoned)

(Application Serial No.)

(Filing Date)

(Status)
(patented,pending,abandoned)

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punished by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

POWER OF ATTORNEY: As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith:

Kurt G. Briscoe, Reg. No. 33,141; William C. Gerstenzang, Reg. No. 27,552; Lorimer P. Brooks, Reg. No. 15,155; Bruce Londa, Reg. No. 33,531; Christa Hildebrand, Reg. No. 34,953; and Howard C. Lee, Reg. No. 48,104 all of 220 East 42nd Street, 30th Floor, New York, New York 10017; William R. Robinson, Reg. No. 27,224 of 721 Route 202-206 Bridgewater, New Jersey 08807; Davy E. Zonerach, Reg. No. 37,267, Mark A. Montana, Reg. No. 44,948 and Robert A. Hyde, Reg. No. 46,354, of 721 Route 202-206, Bridgewater, New Jersey 08807, my attorneys with full power of substitution and revocation.

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FULL NAME OF FOURTH INVENTOR:

INVENTOR'S SIGNATURE:

DATE:

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FULL NAME OF FIFTH INVENTOR:

INVENTOR'S SIGNATURE:

DATE:

RESIDENCE:

CITIZENSHIP:

POST OFFICE ADDRESS:

FULL NAME OF SIXTH INVENTOR:

INVENTOR'S SIGNATURE:

DATE:

RESIDENCE:

CITIZENSHIP:

POST OFFICE ADDRESS:

FULL NAME OF SEVENTH INVENTOR:

INVENTOR'S SIGNATURE:

DATE:

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